

# Millimeter Wave Surface Resistance of $\text{RBa}_2\text{Cu}_3\text{O}_{7-\delta}$ ( $\text{R} = \text{Y}, \text{Eu}, \text{Dy}, \text{Sm}, \text{Er}$ ) Superconductors

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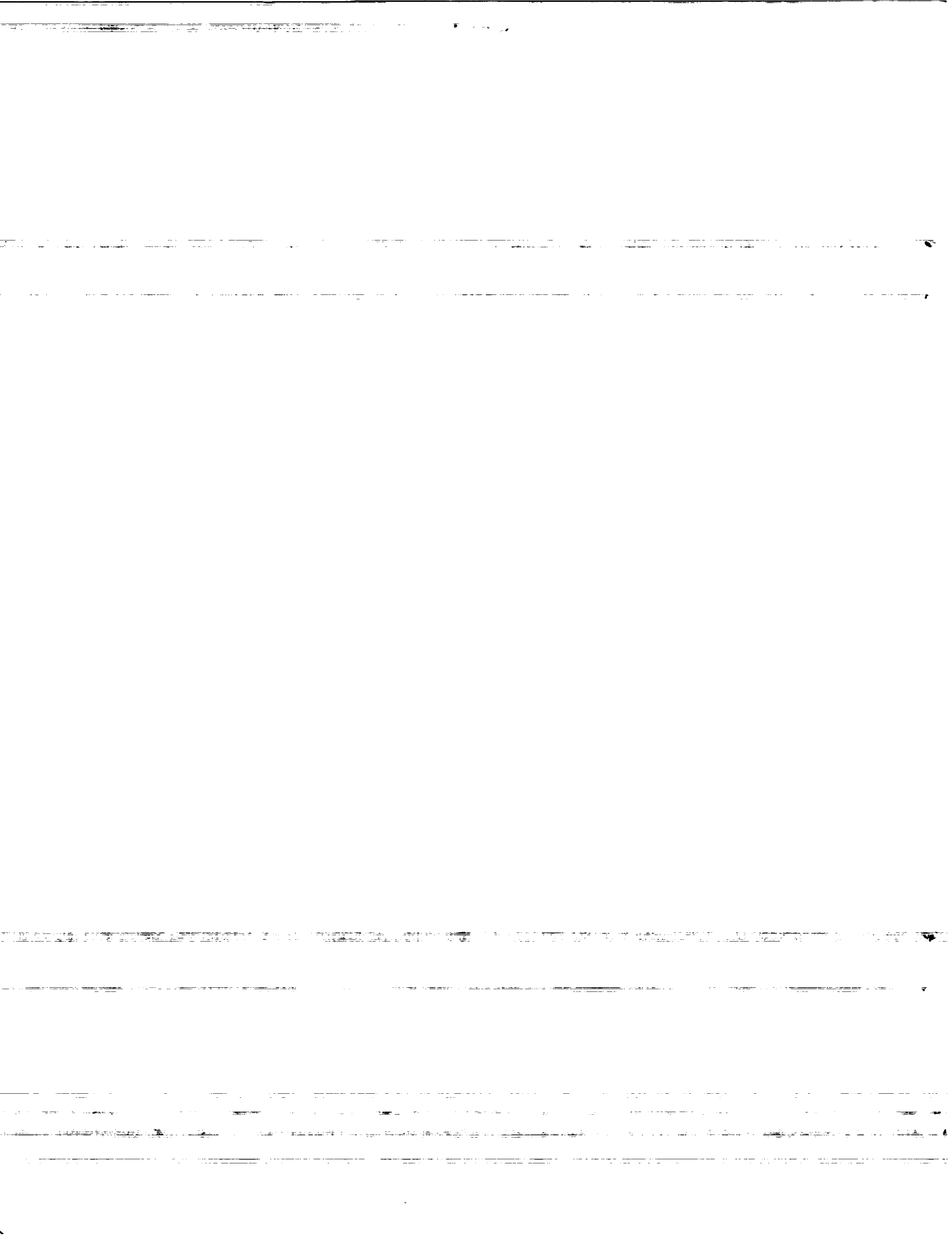
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# MILLIMETER WAVE SURFACE RESISTANCE OF $\text{RBa}_2\text{Cu}_3\text{O}_{7-\delta}$

( $R = \text{Y, Eu, Dy, Sm, Er}$ ) SUPERCONDUCTORS

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## SUMMARY

We report on the measurements of the millimeter wave surface resistance ( $R_s$ ) at 58.6 GHz of bulk samples of  $\text{RBa}_2\text{Cu}_3\text{O}_{7-\delta}$  ( $R = \text{Y, Eu, Dy, Sm, Er}$ ) and of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  superconducting films, in the temperature range from 20 to 300 K. The bulk samples were prepared by cold pressing the powders of  $\text{RBa}_2\text{Cu}_3\text{O}_{7-\delta}$  into 1 in. diameter disks which were sintered at 925 °C in one atmosphere of oxygen. The thin films were deposited on  $\text{SrTiO}_3$  and  $\text{LaGaO}_3$  substrates by pulsed laser ablation. Each sample was measured by replacing the end wall of a gold-plated  $\text{TE}_{013}$  circular mode copper cavity with the sample and determining the cavity quality factor  $Q$ . From the difference in the  $Q$ -factor of the cavity, with and without the sample, the  $R_s$  of the sample was determined.

## INTRODUCTION

Investigation of the properties of the high transition temperature ( $T_c$ ) superconductors at millimeter wave frequencies is important not only to evaluate their potential for practical microwave applications but also in an attempt to determine the extent to which the standard microscopic theories are able to describe the phenomena of superconductivity in these new materials. From the application point of view, the main interest is to determine how well these materials will perform when implemented to transmission lines and microwave devices in comparison with the most commonly used normal metals (Cu and Au) and lower  $T_c$  superconductors currently in use. A parameter which directly provides this information is the surface resistance ( $R_s$ ). To date, a considerable amount of work has been done on measurements of the  $R_s$  both in bulk and thin film high  $T_c$  superconductors at different temperatures and frequencies (refs. 1 to 6). Nevertheless, to the best of our knowledge, no measurements of  $R_s$  have been reported at frequencies around 60 GHz. In this paper, we report on the measurements of the surface resistance of R-Ba-Cu-O ( $R = \text{Y, Eu, Dy, Sm, Er}$ ) bulk superconductors and  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  superconducting thin films at 58.6 GHz and at temperatures from 20 to 300 K.

## EXPERIMENTAL

The bulk samples were made from cold pressed (20,000 psi) sintered powders made from starting powders of  $\text{BaO}_2$  (99.89 percent pure),  $\text{CuO}$  (99.99 percent pure) and  $\text{R}_2\text{O}_3$  ( $\text{R} = \text{Y, Eu, Dy, Sm, Er}$ ), (99.99 percent pure). The powders were ground together by hand, fired at 925 °C in flowing oxygen ( $\text{O}_2$ , 99.995 percent pure) for 6 hr, and were cooled to 450 °C at the rate of 2 °C/min. The material was then held at 450 °C for 6 hr before being cooled to room temperature at 2 °C/min. Afterwards, the powders were reground and refired using the same procedure. After the second firing x-ray diffraction showed that the powders were in the superconducting phase. Then the powders were ground and pressed into disks of 1 in. diameter and 1/4-in. thickness and fired at 925 °C.

The pulsed laser ablation technique used for the deposition of the film is similar to that used by other researchers (refs. 7 and 8). The deposition was performed at a substrate temperature of 750 °C at an ambient oxygen pressure of 170 mtorr. The laser wavelength was 248 nm, the pulse length was 20 to 30 ns, and the pulse rate was 4 pps. During deposition, the distance between the target and the sample was kept at 7.5 cm and the laser fluence on the target was maintained at 2.0 J/cm<sup>2</sup> per pulse. During this process, the laser beam was scanned up and down 1 cm over the target using an external lens on a translator. At the end of the deposition process, the oxygen pressure was raised to 1 atm, and the temperature was lowered to 450 °C at a rate of 2 °C/min. The temperature was held at 450 °C for 2 hr before it was lowered to 250 °C at a rate of 2 °C/min. The heater power was turned off and the sample was allowed to cool down to 40 °C or less before it was removed from the chamber. A more detailed description of the deposition technique is given in reference 9.

Surface resistance measurements of both types of samples were made at 58.6 GHz applying the same experimental technique. Using an HP-8510 network analyzer and Ginzton's impedance method (refs. 10 and 11) the Q-factor of the cavity was determined from the reflection coefficient. In each case the end wall of the cylindrical cavity ( $\text{TE}_{013}$  mode) was replaced by the superconducting sample, and  $R_s$  was calculated from the difference in Q values of the bare cavity and the cavity with the sample in place. All the measurements were taken at temperatures from 20 to 300 K, and under a vacuum of less than 10 mtorr.

## RESULTS

The dc resistance versus temperature measurements were performed using a standard four probe method. For the bulk samples the transition temperatures ( $T_c$ ,  $R_{dc} = 0$ ) were distributed between 91.8 K (Eu-Ba-Cu-O) to 79.0 K (Y-Ba-Cu-O and Sm-Ba-Cu-O), as can be seen from dc resistance versus temperature curves shown in figures 1(a) and (b). All the samples had densities from 50 to 60 percent of the ideal, and from SEM micrographs grain sizes of approximately 5  $\mu\text{m}$  were observed. Figure 2 exhibits the measured Q-factor for the cavity as a function of temperature for cases in which its end wall had been replaced by each of the bulk samples under study. It is observed that the Y, Eu, and Dy based samples show a clear increase of the Q-factor at temperatures below  $T_c$ , while a rather discrete change is noticed for the Sm-based sample. No change in the rate of increase of Q with decreasing temperature, at temperatures below  $T_c$ , is observed for the Er-Ba-Cu-O sample.

Therefore, these observations appear to suggest that although the cold pressing method yields bulk samples with reasonably good  $T_C$  values, it does not guarantee obtaining samples with low  $R_S$  values. Clear evidence of this is observed in particular in the Er-based sample, and to less extent in the Eu-based sample. The behavior of the Q-factors for the different samples considered is in fair agreement with the concept that the losses in these ceramics appear to be dominated by the weak coupling between grains (ref. 12), especially for grains close to the surface where most of the losses take place.

Figure 3 shows the experimental values of the surface resistance for the Y-, Dy- and Eu based bulk samples. Also plotted is the experimental surface resistance for the gold-plated copper cavity for comparison. It is observed that the  $R_S$  for the  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  sample is smaller than that for the other bulk samples, at temperatures below  $T_C$ . The  $R_S$  values for the samples at temperatures below  $T_C$  are beyond a factor of 2 smaller than the values in the normal state. Nevertheless, none of the samples showed an  $R_S$  better than that of the gold-plated cavity even at temperatures below  $T_C$ . The fact that the surface resistance obtained for these samples is not comparable with that of gold at any temperature is evidence of the poor quality of the surface of samples prepared by the preparation process considered in this study.

Figure 4 shows the dc resistance versus temperature curves corresponding to  $\text{YBa}_2\text{Cu}_2\text{O}_{7-\delta}$  superconducting films deposited on  $\text{SrTiO}_3$  and  $\text{LaGaO}_3$  substrates by laser ablation. Zero dc resistance was attained at 90.0 and 88.9 K for the films on  $\text{SrTiO}_3$  and  $\text{LaGaO}_3$ , respectively. The x-ray diffraction pattern revealed that both films are predominantly c-axis oriented, while SEM micrographs showed that both films are polycrystalline, as can be seen from figure 5. Figure 6 shows the measured Q-factor for the cavity as a function of temperature for cases in which its end wall had been replaced by the  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  films. A clear increase in the Q-factor is observed for both films at temperatures below  $T_C$ , with the rate of increase of the Q-factor with decreasing temperature being higher for the film on  $\text{SrTiO}_3$  than for the one on  $\text{LaGaO}_3$  down to 50 K. At temperatures below 50 K we are limited by the resolution of our measurements. Therefore, contrary to what was observed in the bulk material, the value of  $T_C$  appears to be directly related with lower microwave losses.

The measured  $R_S(T)$  curves for the two films under study are shown in figure 6, in addition to the curve corresponding to the gold-plated copper cavity. The  $R_S$  of the films is comparable in the normal state, while the  $R_S$  for the film on  $\text{SrTiO}_3$  was lower than that for the film on  $\text{LaGaO}_3$  at temperatures just below  $T_C$ . Using the normal skin depth formula  $R_S = (\omega\mu_0\rho/2)^{1/2}$  a typical resistivity  $\rho$  at 300 K of approximately 118 and 158  $\mu\Omega\text{-cm}$  is obtained for the film  $\text{SrTiO}_3$  and  $\text{LaGaO}_3$ , respectively. In the superconducting state the films on  $\text{SrTiO}_3$  and  $\text{LaGaO}_3$  exhibit a drop of  $R_S$  to effective values of 103 and 144 m $\Omega$  at 77 K, and 82 and 116 m $\Omega$  at 70 K, respectively. The surface resistance at 77 K for the film on  $\text{SrTiO}_3$  is less than that of the gold-plated cavity, while for the film on  $\text{LaGaO}_3$   $R_S$  is the same as for the gold-plated cavity. Nevertheless, the value of  $R_S$  at 77 K for both films is higher than the theoretical  $R_S$  value expected for copper at the same temperature and frequency.

Since we are operating at a fixed frequency, we cannot study the frequency dependence of  $R_S$  directly from our measurements. Nevertheless, a comparison

of the  $R_s$  values with those reported by other researchers in similar types of films and at different frequencies, may be helpful to formulate a frequency dependence trend for  $R_s$ . Thus, using the results obtained by Klein, et al. (ref. 13) for c-axis textured layers samples of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ , and fitting their data to a quadratic frequency dependence for  $R_s$ , gives an  $R_s$  value of 102 m $\Omega$  at 58.6 GHz and 77 K. This value agrees very well with our experimentally obtained value of 103 m $\Omega$  at the same temperature. A similar approach was used for the film on  $\text{LaGaO}_3$ . Using the values for  $R_s$  at 22, 86, and 148 GHz and at 70 K reported by Cooke, et al. (ref. 14) for a  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  superconducting film deposited by magnetron sputtering on  $\text{LaGaO}_3$ , we were able to find by interpolation an  $R_s$  value of 106 m $\Omega$  at 58.6 GHz. This value is within experimental uncertainty of our measured value of 116 m $\Omega$ . These results indicate that our value fit well with the nearly quadratic dependence for  $R_s$  ( $R_s \propto \omega^n$ ,  $n = 2.06 \pm 0.14$ ) reported by Cooke, et al. (ref. 14). Our results, as well as those obtained by the above mentioned researchers, indicates that the quadratic dependence observed for  $R_s$  is consistent with the experimental behavior observed in low  $T_c$  superconductors and also with the predictions of the BCS theory.

## CONCLUSIONS

There appears to be no direct correlation between the surface resistance  $R_s$  and the transition temperature  $T_c$  in the  $\text{RBa}_2\text{Cu}_3\text{O}_{7-\delta}$  bulk superconducting samples obtained by the cold pressing preparation method. Therefore, the dominating factors which control the microwave losses can only be guessed at. Some of the possible factors which could control the losses are: the weak coupling at grain boundaries, the purity of the sample and the possible segregation of composition at the surface. Two main factors can contribute to the presence of weak coupling. The first is the intrinsic mismatch of the lattice and the segregation of impurities to the grain boundaries, while the second could be reactions with the ambient environment (i.e., water vapor and  $\text{CO}_2$ ). This environmental reactions primarily occur at the surface and would not appreciably affect the bulk properties in a short time period. If the losses are due to either the reaction of the surface with the environment or due to porosity, then a different pressing process to make denser pellets should give samples with lower microwave losses.

For the  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  superconducting films on  $\text{SrTiO}_3$  and  $\text{LaGaO}_3$  we found from a correlation of the  $R_s$  values obtained for both films with those obtained for similar films measured by other researchers at different frequencies, that these values are consistent with the frequency dependence for  $R_s$  observed in classical superconductors, and also with the predictions of the BCS theory.

In summary, we have measured the  $R_s$  of bulk  $\text{RBa}_2\text{Cu}_3\text{O}_{7-\delta}$  ( $R = \text{Y, Dy, Eu, Sm, Er}$ ) superconducting samples and found that none of them performed as well as gold. Also, for the 1.2  $\mu\text{m}$  thin films of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  on  $\text{SrTiO}_3$  and  $\text{LaGaO}_3$  we obtained values of  $R_s$  significantly lower than gold at temperatures below 70 K, but we are limited by the resolution of our measurement in accurately determining  $R_s$  values below 50 K.

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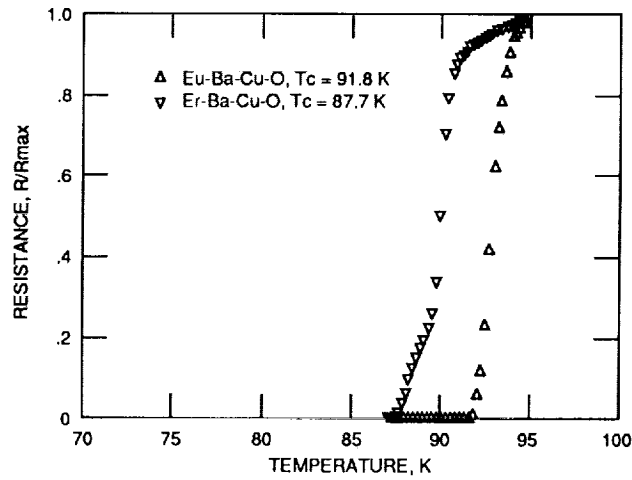
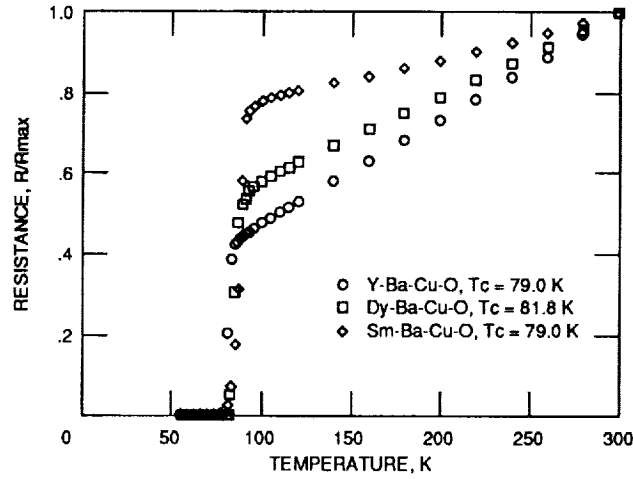


Figure 1. - dc resistance versus temperature measurements of bulk superconducting samples.

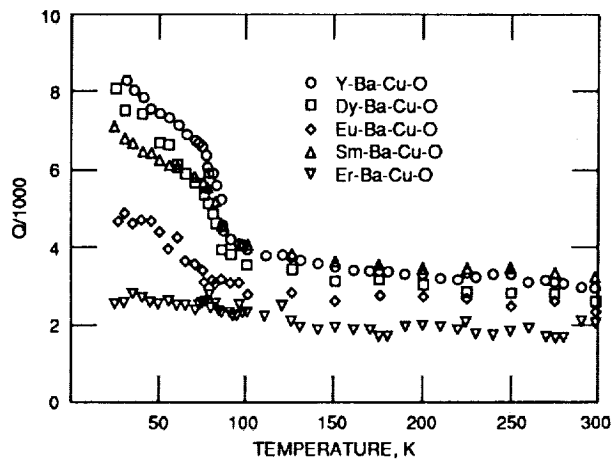


Figure 2. - Measured  $Q$  of the cavity, with one end wall replaced by the bulk sample, as a function of temperature.

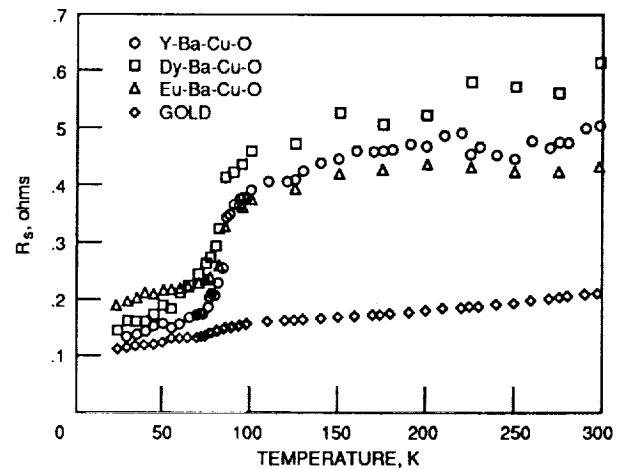
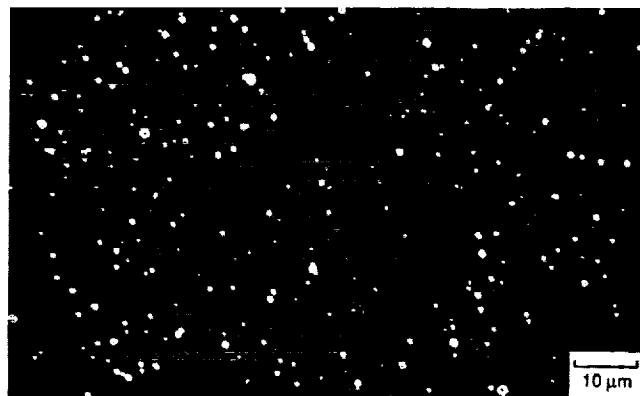


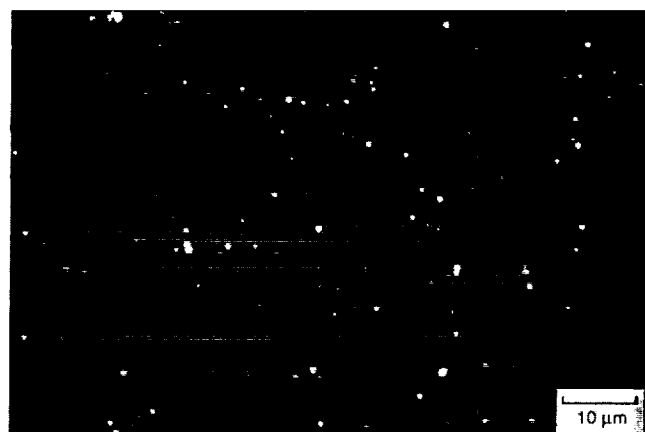
Figure 3. - Surface resistance  $R_s$  as a function of temperature and at 58.6 GHz, for bulk superconducting samples.



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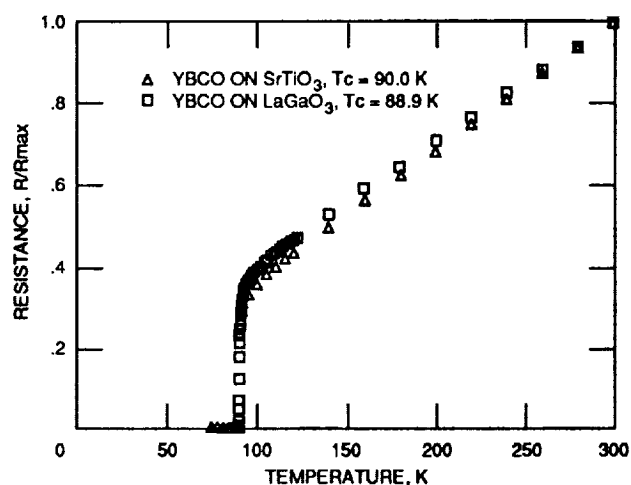


Figure 4. - dc resistance versus temperature measurements of laser ablated  $\text{YBa}_2\text{Cu}_3\text{O}_{7.8}$  superconducting films on  $\text{SrTiO}_3$  and  $\text{LaGaO}_3$  substrates.

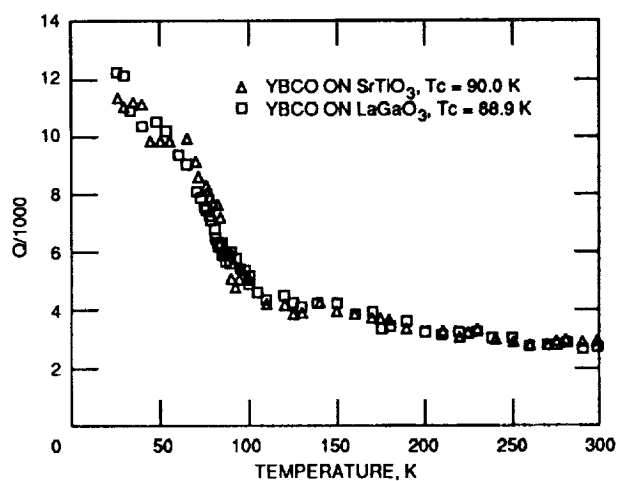


Figure 6. - Measured  $Q$  of the cavity, with one end wall replaced by the thin film, as a function of temperature.

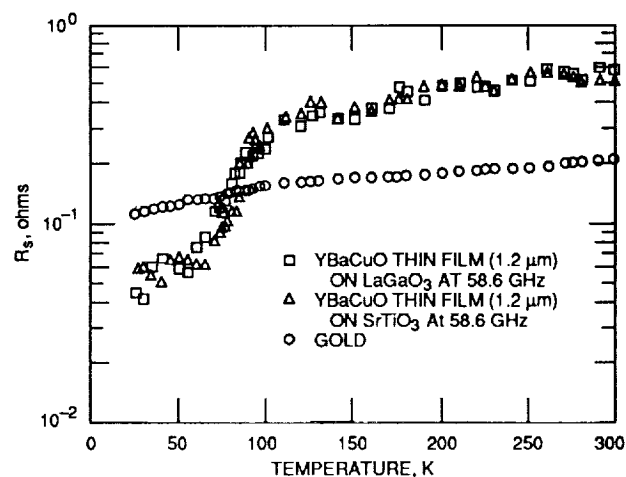


Figure 7. - Surface resistance ( $R_s$ ) at 58.6 GHz versus temperature for 1.2  $\mu\text{m}$  films of  $\text{YBa}_2\text{Cu}_3\text{O}_{7.8}$  deposited by laser ablation onto  $\text{SrTiO}_3$  and  $\text{LaGaO}_3$  substrates, and for gold-plated cavity.

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